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In re Patent Application of:

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For: THERMOELECTRIC SEMICONDUCTOR MATERIAL, THERMOELECTRIC SEMICONDUCTOR ELEMENT THEREFROM, THERMOELECTRIC MODULE INCLUDING THERMOELECTRIC SEMICONDUCTOR ELEMENT AND PROCESS FOR PRODUCING THESE

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DECLARATION AUTHENTICATING THE ENGLISH-LANGUAGE TRANSLATION OF PORTIONS OF JAPANESE PATENT DOCUMENT

I, Naoki Fukai, declare under penalty of perjury that I understand well the Japanese language and the English language, that, to the best of my knowledge and belief, the statements in the English language in the attached translation of portions of Horio et al., JP 2003-037302 A have the same meanings as the statements in the Japanese language in the original document, a copy of which I have examined, and that I am the person whose signature appears below.

Name: Naoki Fukai

Date: September 30, 2011

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English-language Translations

[0029]

Thereafter, the ribbon 14 is crushed as necessary and classified so as to make the particle size uniform. In addition, an appropriate particle size range of the ribbon (foil) 14 is laminated and charged in a prismatic die (not shown) and hot-pressed by applying a pressure P while restricting the side face with the heated hot pressing. Accordingly, as shown in FIG. 7, a prismatic solidified compact 61 is obtained (primarily solidification-molded), which has a crystal structure having a crystal grain where the long axes are made uniform in the pressing direction (pressurizing direction) and the short axes are made uniform in a direction orthogonal to the pressurizing direction (Step S6).

[0030]

Deformation and flaws are introduced into quenching slices manufactured by the liquid quenching technique. When solidification-molding the quenching slices by hot-pressing or the like with or without crushing the quenching slices, the crystal grain is grown or a recrystallized grain is precipitated with the deformation and the flaws as a crystal nucleus. This recrystallized grain can be made to be a crystal grain which has a large aspect ratio having the long axis in a direction parallel to the pressing direction during the solidification molding and having the short axis orthogonal to the pressing direction.

[0031]

Therefore, when the quenching slices (powder) are solidification-molded, the quenching slices are pressurized in a direction parallel to the thickness direction of the quenching ribbon, that is, in a direction parallel to the long axis of the crystal grain in the quenching ribbon and the long axes of the recrystallized grain generated during the solidification molding are made uniform in the pressurizing direction. As a result, a solidified compact is obtained, which has a crystal structure where the long axial direction of the crystal grain is made uniform in a direction parallel to the pressurizing direction.

[0032]

In addition, in the present embodiment, as described above, since Te atoms and Se atoms are segregated on the surface of the quenching ribbon 14 by the thermal treatment, these atoms are easily diffused to each other between the quenching ribbons 14 and easily solidification-molded.

[0033]

Thereafter, the side-face pressing (Step S7a), the upset forging (Step S7b), the

rolling process (Step S7c) or the hot isostatic pressing (Step S7d) is performed on the solidified compact.

[0034]

FIG. 8 is a diagram schematically showing the method of the side-face pressing. When performing the side-face pressing, the solidified compact 61 is warm-pressed or hot-pressed from a first or second direction parallel to the pressurizing direction of the hot pressing during the primary solidification molding. For example, in FIG. 8, when the pressure during the hot pressing is represented by P1, the pressure represented by P2 and/or P3 is applied to the solidified compact.

[0035]

FIG. 9 is a diagram schematically showing the upset forging method. When performing the upset forging, first, the solidified compact 61 is rotated by 90° so as to be in a horizontal direction of the pressurizing direction of the hot-pressing as shown in FIG. 9(a) and is pinched between flat dies 62 as shown in FIG. 9(b) to apply the pressure (P4) thereto. As a result, as shown in FIG. 9(c), each crystal grain is further extended in the pressurizing direction of the hot pressing, thereby improving the orientation. Here, the pressure is, for example, 0.5 to 2.0 t/cm^2 and the temperature of the solidified compact 61 at the time is maintained at, for example, 350 to 500°C.

[0048]

In Examples 14 to 19, slices or powders were prepared from ingots blended into each composition according to the liquid quenching technique, followed by annealing in the atmosphere of Ar at 400°C for 10 hours, laminating the foil, and hot pressing in the thickness direction of the foil (a direction parallel to C plane) as the pressurizing direction, thereby preparing the solidified compact. Furthermore, the solidified compact was vacuum-sealed into a sheath made of aluminum to perform the sheath rolling. During the hot pressing, a cemented carbide die was used in the atmosphere of AR. In addition, the shape of the solidified compact was a parallelepiped of which the length of each side was 50 mm. To P-type thermoelectric materials (Examples 14 to 16), the pressure of 0.5 (t/cm^2) was continuously applied at 350°C for 1 hour. To N-type thermoelectric materials (Examples 17 to 19), the pressure of 0.5 (t/cm^2) was continuously applied at 400°C for 1 hour. Further, during the sheath rolling, the solidified compact was cut into half of which the shape was a parallelepiped with two sides of the length of 50 mm and the other one side of the length of 25 mm, followed by rolling under the conditions that the rolling temperature was 450°C, the roll diameter was 400 mm, the

rolling speed was 2 m/sec, and the rolling reduction was 20 %.

FIG. 1

FIG. 1 is a flowchart showing a method of manufacturing a thermoelectric material according to an embodiment of the present invention.

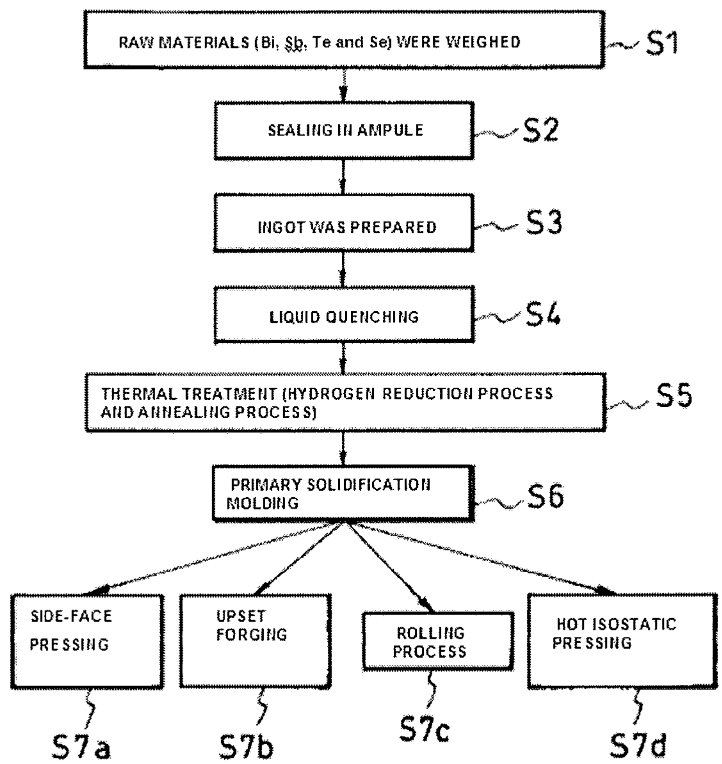

FIG. 3

FIG. 3 is a diagram showing a method of manufacturing a powder of a thermoelectric material according to the liquid quenching technique.

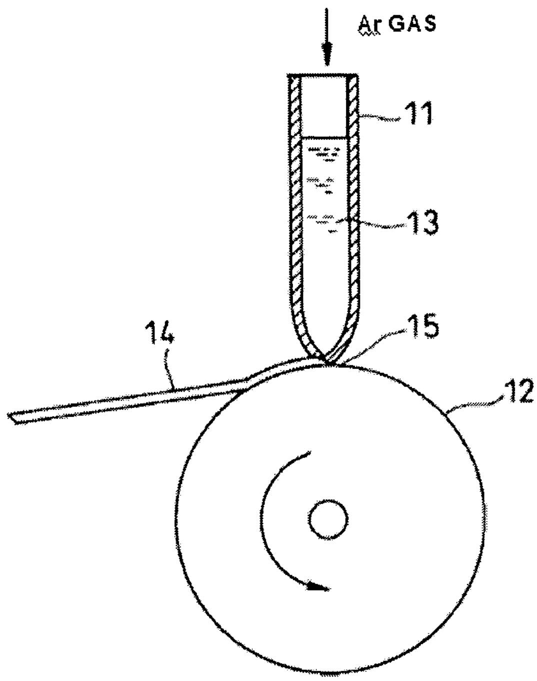

FIG. 7

FIG. 7 is a diagram showing a solidification molding method using a hot pressing.

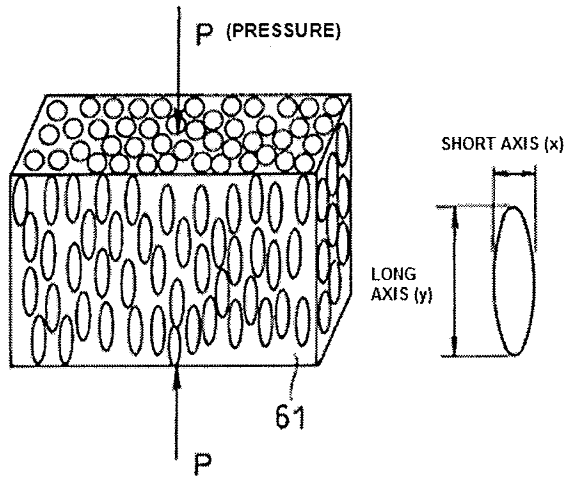

FIG. 8

FIG. 8 is a diagram schematically showing a side-face pressing method.

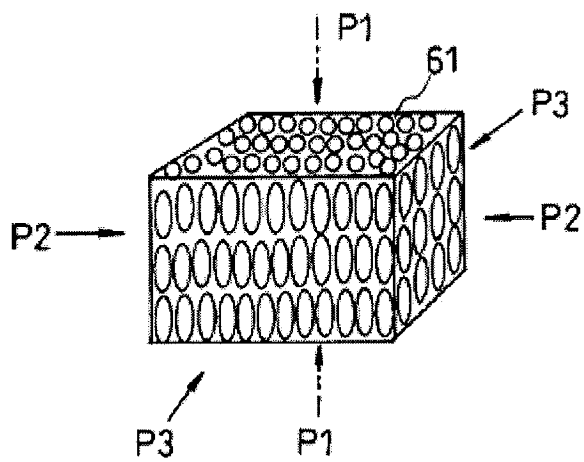

FIG. 9

FIG. 9 is a diagram schematically showing an upset forging method.

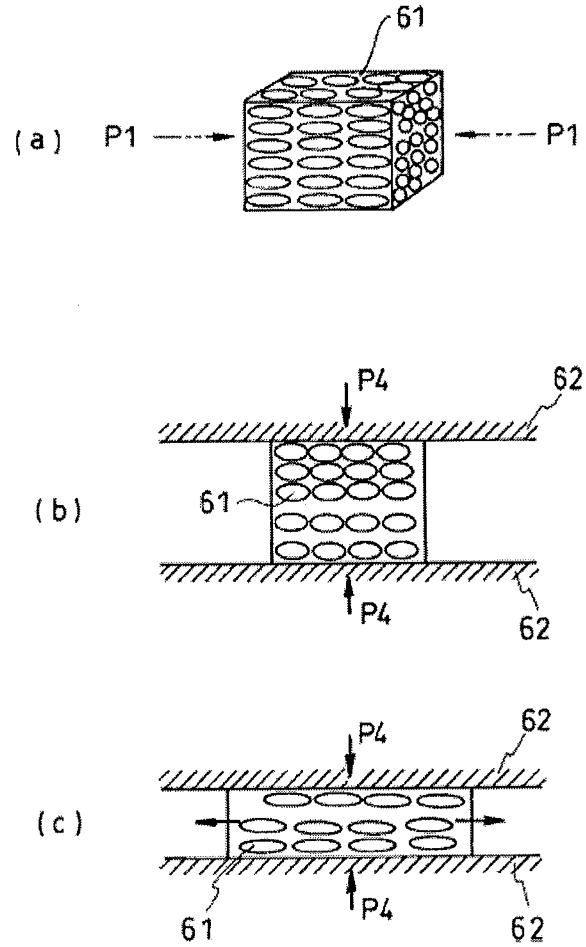

FIG. 10

FIG. 10 is a diagram schematically showing a sheath rolling method as an example of a rolling process.